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16. ABSTRACT

To determine the nature of aerosol particles in a rural area of the eastern United States, aerosol samples were collected at Deep Creek Lake, Maryland, on various substrates and analyzed by a scanning electron microscope (SEM) and a transmission electron microscope (TEM). SEM analysis of particles larger than 2.5 μm collected on Nuclepore filters revealed the following: clay minerals, quartz, gypsum, and calcite comprised 50% of the particles analyzed; spores, pollen, and plant debris comprised 25%; 9% were fly ash; and 11% were sulfates. Particles ranging from 0.3 to 2 μm were collected in a Cascade impactor on grid-supported carbon films and analyzed by TEM for decomposition rate as well as for reaction with the barium chloride and nitron films that were applied. The TEM analyses indicated that as much as 95% of the particles in the 0.3- to 2-μm diameter range were pure ammonium sulfate or acidic ammonium sulfate; they contained essentially no insoluble or nonvolatile matter. About 5% of the particles were fly ash spheres. Replicas of particles collected on Nuclepore filters were analyzed by TEM; this revealed that chain agglomerates of elemental carbon were abundant in particles smaller than 0.1 μm.

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Characteristics of Individual Particles at a Rural Site in the Eastern United States

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To determine the nature of aerosol particles in a rural area of the eastern United States, aerosol samples were collected at Deep Creek Lake, Maryland, on various substrates and analyzed by a scanning electron microscope (SEM) and a transmission electron microscope (TEM). SEM analysis of particles larger than 2.5 μm collected on Nuclepore filters revealed the following: clay minerals, quartz, gypsum, and calcite comprised 50 percent of the particles analyzed; spores, pollen, and plant debris comprised 25 percent; 9 percent were fly ash; 11 percent were sulfates; 5 percent were unidentified. Particles ranging from 0.3 to 2 μm were collected in a cascade impactor on grid-supported carbon films and analyzed by TEM for decomposition rate as well as for reaction with the barium chloride and nitron ($C_{20}H_{18}N_4$) films that were applied after sampling. The TEM analyses indicated that as much as 95 percent of the particles in the 0.3- to 2-μm diameter range were pure ammonium sulfate or acidic ammonium sulfate; they contained essentially no insoluble or nonvolatile matter. About 5 percent of the particles were fly ash spheres. When replicas of particles collected on Nucleopore filters were analyzed by TEM, we observed agglomerates of particles smaller than 0.1 μm.

During August and September of 1983 the Allegheny-Deep Creek Lake Experiment was conducted by several research groups to evaluate the feasibility of using receptor models to determine the impact of primary and secondary emissions from fossil fuel combustion sources on ambient aerosol in rural areas. The study consisted of measurements of ambient aerosols at three ground level sites, source sampling at selected coal-fired power plants, and airborne sampling in power plant plumes.[1] The ambient sampling sites were Deep Creek Lake (DCL), Laurel Hill, and Allegheny Mountain, as shown in Figure 1. The sampling site at DCL is located in a rural area of western Maryland (39°34′N, 79°20′W) surrounded by over fifteen coal-fired power plants, which are big enough (>1000 MW) and close enough (50 to 300 km) to have a significant impact on the site.[1] Most of the power plants are located west and northwest of the DCL site, as shown in Figure 1. The study was coordinated by the U.S. Department of Energy, the Electric Power Research Institute, and the U.S. Environmental Protection Agency.[1]

When receptor models[2,3] are applied to data on concentrations of elements in bulk samples, the number of sources that can be resolved is rather limited. The use of electron microscopy to characterize the composition of individual particles may provide direct evidence on source contribution and can increase the resolution of sources in receptor models.[4,5] The main advantage of electron microscopy as an analytical tool is in its ability to analyze individual particles (rather than bulk) to determine their composition, heterogeneity, and surface features, all of which provide clues on particle source, transport, and transformation. The application of electron microscopy to atmospheric aerosols requires very short sampling times to avoid particle pile-up; spacing of at least three diameters is desirable. Although the need to have short sampling periods complicates the task of obtaining representative samples, the ability of electron microscopy to reveal the nature of particles makes that technique a valuable complement to bulk sample analysis. We have used a scanning electron microscope (SEM) and a transmission electron microscope (TEM) mainly for qualitative analysis. Although it is also possible to sum the properties of individual particles to obtain a quantitative analysis of mass and elemental concentrations, such an analysis is based on assumptions of density and shape that can lead to large inaccuracies. For example, particle dimensions and density are needed to calculate mass concentration. In the coarse fraction at DCL, particles had irregular shapes and unknown thicknesses; some were loose aggregates of unknown density.[6] Submicrometer-sized particles, especially sulfates, nitrates, and hydrocarbons, may change their size under the electron beam and the high vacuum.

The purpose of this paper is to describe the application of electron microscopy to the analysis of individual particles collected at DCL and to report the particles' main features. In our study, an SEM coupled to an energy dispersive x-ray (EDX) spectrometer was used to characterize particles larger than 0.5 μm collected on Nuclepore filters. A TEM was used to characterize the morphology of 0.05- to 5-μm particles collected on grids which were chemically treated to reveal the nature of the particles. The TEM also was used to analyze replicas of particles collected simultaneously on Nuclepore filters. Results are reported elsewhere for SEM analysis of large particles (>5 μm) collected on nylon films by a rotary inertial impactor at DCL.[6]

Experimental Procedure

In the present work, particles were collected and analyzed by electron microscopy using the techniques described below.

SEM Analysis of Particles on Nuclepore Filters

Aerosol samples were collected on 0.4-μm pore diameter Nuclepore filters for 1–6 h. Each filter was later coated under vacuum with a thin film of carbon (5–10 nm) and analyzed by SEM and EDX. Random fields of view were chosen, normally 5–10, and several hundred particles were characterized for each case. The results include size, morphology, and elemental content of the individual particles. EDX spectra

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were obtained using 30-s counting periods, a 30-kV electron beam having 80 to 100 mA current, and an EDX detector 12 mm from the sample. A Be window on the EDX detector limited the analysis to elements having atomic numbers ≥11. Spectral analysis was performed by using a simple computer program for peak identification. In case of overlapping peaks, all elements that could be attributed to the peaks were listed by the computer program, and the operator made the appropriate identification based on peak shapes and related spectral lines. The size of each particle was manually recorded, and specific features (shape, morphology, and category) were noted. During analysis by SEM-EDX, each particle was assigned to a defined category based on its shape, surface properties, and elemental content. Particles of biological origin such as spores, pollen and plant debris were identified by a high background in their x-ray spectra and a lack of abundant elements with atomic numbers above 11. Mineral particles were identified on the basis of an irregular shape and high abundance of elements as follows: quartz (Si), dolomite (Mg, Ca), gypsum (S, Ca), and clay (Mg, Al, Si, K, Ca, Fe). Coal fly ash was identified by the same elements and a spherical shape. In this mode, particles larger than 0.5 μm were analyzed. Although smaller particles could be analyzed with an SEM-EDX system, we used the TEM techniques described below to characterize particles as small as 0.05 μm.

TEM Analysis of Particles on Carbon Films

While samples were collected on Nuclepore filters for SEM analysis, particles for TEM analysis were also collected on carbon films that were supported by 3-mm diameter copper grids placed below the jet nozzle of each stage of a Casella cascade impactor.[7] The impactor stages (and the ranges of diameters observed for the collected particles) were as follows: Stage 4 (0.3 to 1 μm), Stage 3 (0.8 to 2 μm), and Stage 2 (>2 μm). The overlap in size ranges for Stages 3 and 4 results in part from a lack of steepness in the impactor's fractionation characteristics. Air was sampled at a rate of 15 to 20 L/min^{-1} at a height of 2 m above the ground in an open field. Sampling times were kept very short (10 to 100 s) for Stage 4 and sufficiently long (10 to 100 min) for larger particles collected on Stages 3 and 2. With such short sampling times, particles do not pile up, and artifact formation is expected to be minimal.

On each stage, particles were simultaneously collected on a set of four grids. One grid that received no further treatment was analyzed by TEM to obtain particle size, shape, and number concentration. Also, particles' rates of decomposition under the 100 kV electron beam were observed qualitatively at magnifications of 3,000 to 10,000. The rate and completeness of particle decomposition provided clues on particle volatility, homogeneity, and composition. After the samples were returned to the laboratory, the second grid was coated under vacuum with a thin film of barium chloride ($BaCl_2$). Particles containing sulfate reacted with the film and formed specific reaction spots recognizable in the TEM. The reaction spots of sulfuric acid (H_2SO_4) particles and acidic ammonium sulfate are similar, but they are different both from that of ammonium sulfate [$(NH_4)_2SO_4$] and from that of insoluble material coated with sulfate.[8] After sampling, the third grid was coated under vacuum with a thin film of nitron ($C_{20}H_{16}N_4$) to identify nitrate particles.[9] The nitrate identification is based on the appearance of fibers that appear after reaction with nitron.[9] Ammonium nitrate not treated with nitron is very unstable under the TEM beam but is less affected by the vacuum in the coating process.[9] When the nitron film reacted with sulfates, characteristically different reaction spots appeared, which provided information on individual sulfate particles. The fourth grid was kept as a spare sample, or in some cases was coated with nitron before sampling in order to study volatile nitrate. Some of the grids were also analyzed by SEM-EDX to identify particles by both morphology and elemental composition.

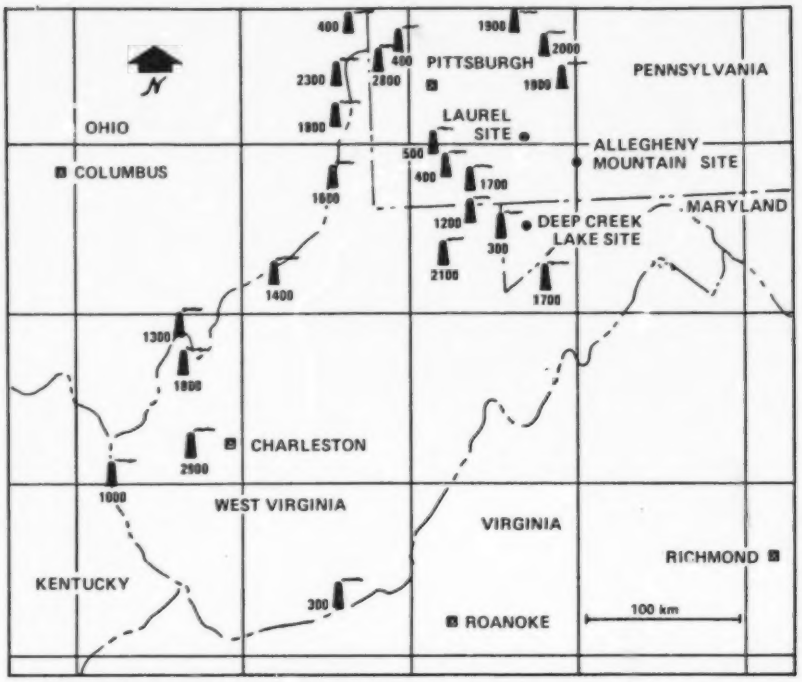

Figure 1. The Deep Creek Lake, Laurel Hill, and Allegheny Mountain sampling sites in relation to major power plants whose output is shown in megawatts (based on data from Stevens[1]).

TEM Analysis of Replicas of Particles

Ultra-fine particles (<0.1 μm) were too small to be analyzed for elemental content by the SEM-EDX system. These particles, which are collected efficiently on Nuclepore filters,[10] were transferred onto an electron microscope grid by preparing replicas that could be studied in the TEM without the danger of particle evaporation. To prepare a replica, a 3-mm × 3-mm section of the Nuclepore filter was coated under vacuum with a 20-nm thick carbon film. The coated filter was then placed on a 3-mm copper grid in a closed chamber containing chloroform vapor, which dissolved the Nuclepore filter material within 24 to 48 h and left the replicated particles on the grid.[11] Although the chloroform vapor may dissolve some of the particles, the carbon coating preserves their morphology. Thus, chain agglomerates of ultrafine particles could be observed by this technique.

Results

In this phase of the study several aerosol samples collected on August 16, 19, and 20 (sample Nos. 7, 29, and 32) were analyzed. They provide a preliminary characterization of individual particles in a rural area in eastern United States. When the dichotomous sam-

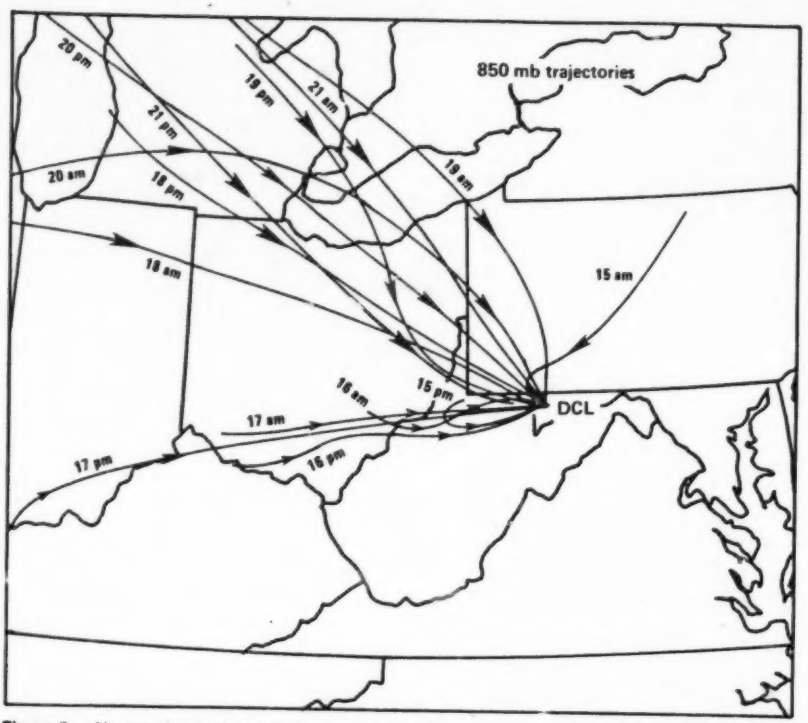

Figure 2. Air parcel trajectories (850 mbar) during the week of sampling for electron microscopic analysis. Date and time of arrival at DCL are shown on each trajectory; arrival at DCL at 0800 and 2000 h is denoted by am and pm, respectively. Arrowheads are spaced at 12-h intervals.

pler data become available (about 3,000 samples were collected and will be analyzed for mass and elemental content[1]), characterization of individual particles will be used to quantify the impact of various sources on ambient aerosol at DCL.

Figure 2 summarizes the 850-mbar air parcel trajectories during the week of the present study.[12] The trajectories were calculated by Samson *et al.*[12] from upper air wind observations taken twice daily at 0800 and 2000 eastern daylight time, at nine stations in the area at a time step of 12 h. Early in the week, the trajectories shifted from northeasterly flow to westerly flow and indicated slow air flow through southern Ohio and the Ohio-West Virginia border. Later in the week (August 18 to 21), the 850-mbar trajectories shifted to a rapid northwesterly flow through Ohio and western Pennsylvania; there was little residence time in the area. Rain was recorded on August 18. Thus, DCL was mainly affected by emissions from power plant located west and northwest of the site during most of the week of August 15 to 21, 1983.

Sulfates

Figures 3A and 3B are typical micrographs of particles collected on Stage 4 (0.3 to 1 μm) of the Casella impactor. Figure 3A shows the particles within the first 10 s of observation under the TEM beam, and Figure 3B shows the same field of view after an additional 30 s of TEM beam exposure. Figure 3 illustrates that exposure to the TEM beam not only causes particles to decompose substantially, but the decomposition is characteristically the same for almost all of the particles. Analysis of this grid enables us to conclude the following:

(a) The particles viewed in the TEM appear spherical.

(b) Most particles (>85 percent) in this size range decompose under the TEM beam in a manner that is characteristic of ammonium sulfate or ammonium bisulfate.[13,14]

(c) About 10 percent of the particles appear darker and have a few satellites around them in a manner typical of particles that are more acidic than ammonium sulfate but are less acidic than sulfuric acid.[13] None resembled pure sulfuric acid particles, which possess a characteristic form: a central droplet surrounded by a large number of much smaller satellites.[14] We cannot exclude the possibility of some neutralization by ambient ammonia occurring after sampling.

(d) The rest (about 5 percent) are solid particles that do not decompose under the TEM beam. Some of these are smooth spheres, which suggests that they are fly ash. Although both oil- and coal-fired power plant fly ash particles in this size range are spherical, other industrial emissions may also possess the same morphology. Our EDX analyses provided direct evidence of the presence of fly ash particles in the samples.

Figure 4 is a typical micrograph obtained by TEM for particles in the 0.8- to 2-μm size range collected on Stage 3 of the impactor. These particles are similar in morphology to those collected on Stage 4, and they evaporated similarly when exposed to the TEM beam. These results were typical of daytime conditions. Particles larger than 1.5 to 2 μm differ completely in shape and composition, as will be shown later. Thus, morphology tells us that about 95 percent of the submicrometer-sized particles are volatile sulfates.

Figure 5 is a typical example of particles collected on Stage 4 (0.3 to 1 μm) and reacted with $BaCl_2$. The reaction spots are indicative of acidic sulfate. Over 95 percent of 509 particles examined appeared to be pure sulfates that did not contain insoluble nuclei. Most of the remaining particles were sulfate with an insoluble core. Essentially the same was true for particles on Stage 3 (0.8 to 2 μm); 90 percent of 569 particles counted were pure sulfates, and only 8 percent were mixed. In most cases, the mixed sulfates consisted of an insoluble solid core, such as fly ash or soot, coated with sulfate. In summary, the $BaCl_2$ reaction spots indicate that over 95 percent of the particles are acidic sulfate.

By combining all the results obtained for Stages 3 and 4 from TEM analyses, one may conclude that most (>95 percent by particle number) of

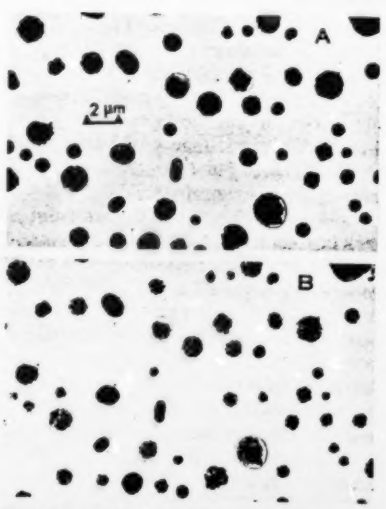

Figure 3. Typical micrographs of 0.3- to 1-μm diameter particles collected on Stage 4 after exposure to the TEM beam: (A) particles after 10-s exposure and (B) the same field of view after 40-s exposure.

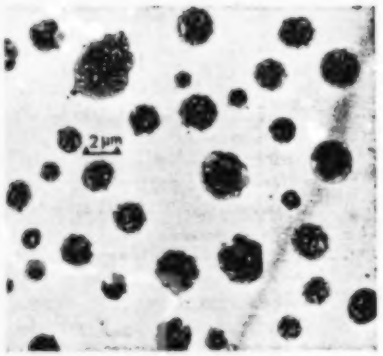

Figure 4. Typical micrograph of particles in the 0.8- to 2-μm range (Stage 3).

the particles in the fine fraction (<2.0 μm) are acidic sulfate, probably ammonium bisulfate. Although the percentages and number of particles quoted refer to Sample 29 (August 19), the conclusion is essentially the same for Samples 7 and 32 (August 16 and 20, respectively), all of which represent daytime conditions. In all cases, several thousand particles were observed in the TEM; however, a smaller number was counted, sized, and observed more closely in the TEM.

The high abundance of acidic sulfate in the particles smaller than 2 μm (Stages 3 and 4 of the impactor) is striking. How do these results based on individual particle analysis compare with those of other studies in which bulk analyses were performed? In an earlier study by Pierson, *et al.* at the Allegheny Mountain site in southwestern Pennsylvania (see Figure 1), the dominant chemical feature of the aerosol was the stoichiometric balance of the $SO_4^=$ anion with NH_4^+ and H^+ cations; H^+ was the more abundant cation when sulfate levels were high.[16] Moreover, sulfate and its associated NH_4^+, H^+, and water constituted nearly all of the aerosol fine mass.[16] Data from rural areas in the Ohio River Valley showed that during extreme events in August 1980, sulfur expressed as ammonium sulfate accounted for 80 percent of fine particle mass (<2.5-μm aerodynamic diameter).[17] On the average during the summer period, sulfur expressed as ammonium sulfate constituted about 60 percent of the fine mass, and the crustal elements plus their related oxides accounted for 3 to 4 percent. The rest, as suggested by Shaw and Paur,[17] were probably carbon compounds and water. In summertime studies in the Great Smoky Mountains and Shenandoah Valley located south of DCL, Stevens *et al.*[18] found that sulfate and related cations accounted for about 60 percent of the fine-fraction mass. Organic carbon accounted for about 2 to 9 percent, elemental carbon accounted for 4 percent, and crustal elements and related oxides accounted for about 2 percent. The average fine particle composition was nearly as acidic as ammonium hydrogen sulfate; several of the samples were somewhat more acidic than ammonium bisulfate.[18] Water bound to the acidic sulfate could account for the remainder. Because water is likely to evaporate in our TEM, and because the other components are low in relative abundance, the above fine particle mass composition is consistent with our fine particle number composition results, which indicate about 5 percent crustal matter and over 90 percent sulfate and related cations.

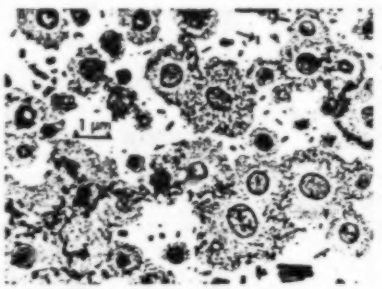

Figure 5. Typical reaction spots of 0.3- to 1-μm particles (Stage 4) with a thin film of $BaCl_2$. The reaction spots are indicative of acidic sulfate. The arrow points to recrystallized $BaCl_2$.

Nitrates

The nitron method was used to detect particles containing nitrate. Figure 6 is a typical example resulting from the application of the technique to particles on Stage 4 of the impactor (0.3 to 1 μm). The reaction spots in Figure 6 are characteristic of sulfates;[9] none of the particles on Stage 4 were nitrates. However, nitrate particles were collected on Stage 3 (0.8 to 2 μm). Figure 7 shows characteristic nitrate reaction spots for two particles. Based on the results of Mamane and Pueschel,[9] we concluded that those spots are characteristic of sodium nitrate and not ammonium nitrate. Some of the nitrates were minerals coated with a nitrate layer. We believe these results are the first direct evidence of the association of nitrate with minerals; however, the number of such mixed nitrate particles was too low to be described in more detail. For Stage 3 (0.8 to 2.0 μm), about 2 to 6 percent of the particles counted contained nitrates (as pure and mixed nitrates).

These results are difficult to compare with bulk nitrate measurements because few studies have differentiated between gaseous nitric acid and particulate nitrate. In the Smoky Mountains study, where differentiation was made, nitrates in the fine fraction (<2.5-μm aerodynamic diameter) accounted for 1

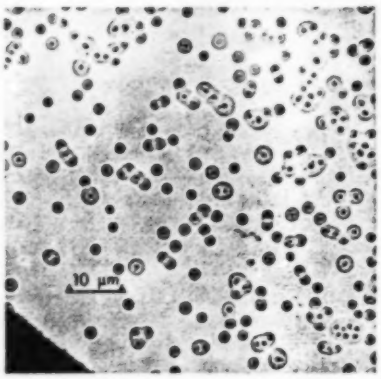

Figure 6. Typical reaction spots of 0.3- to 1-μm particles (Stage 4) with a thin film of nitron. The reaction spots are characteristic of sulfates, not nitrates.

± 1 percent of the mass.[18] If one considers both the existence of a large fraction of mass below 0.8 μm and the possibility that water and organic carbon can evaporate in the TEM, our finding that 2 to 6 percent of the particles in the 0.8- to 2-μm range contained nitrate is not inconsistent with the 1 ± 1 percent nitrates observed in the bulk mass of particles smaller than 2.5 μm.

Coarse Fraction

Although particles larger than 1.5 to 2 μm were collected on Stage 2 of the Casella impactor, their number density was too small for accurate analysis. Instead, particles collected simulta-

Figure 7. Reaction spots of two nitrate particles (impactor Stage 3; 0.8 to 2 μm) with the nitron film. The very few nitrates were found in particles larger than 1 μm. The particle on the bottom left is a "sodium nitrate-type" particle. A reaction of an individual sulfate particle with the nitron film is also seen in the micrograph.

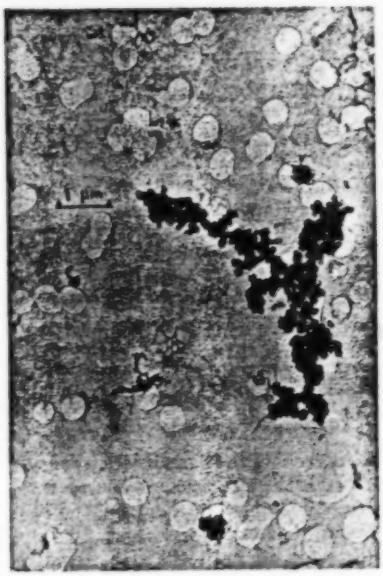

Figure 8. Micrograph of chain agglomerates collected on a Nuclepore filter, as seen in the TEM. Although the filter was dissolved, the 0.4-μm holes are still visible.

neously on Nucleopore filters were analyzed. The result of the analysis shows that particles in the coarse mode belong to three major classes: (a) pollen and plant debris, (b) clay minerals and other minerals such as quartz, calcite, and gypsum, and (c) particles of anthropogenic origin. For Sample 29 (August 19, 1983), 120 particles were individually analyzed; about half were clay and other minerals, about 25 percent were spores, pollen, and plant debris; the rest were of anthropogenic origin—9 percent sulfates, 11 percent fly ash, and a few particles containing metals such as Al, Fe, Zn, and Mn.

Few particles larger than 10 μm were collected on our Nuclepore filters. To collect larger particles, a rotary inertial impactor was used,[19] and SEM-EDX results for individual giant particles are reported elsewhere.[6] Several interesting results have been obtained from the study of the coarse particle mode:[6] (a) Pollen particles form a major portion of the particle population (based on number density). X-ray analysis of the pollen particles indicated a high sulfur content. It has been suggested that part of this sulfur was deposited either as ~0.1-μm sulfate particles (larger ones would have been observed directly in the SEM) or as sulfur dioxide on the surfaces of the pollen particles while they were airborne.[6] (b) Most of the minerals found in the DCL site were clays. Some were of clean crystalline structure, but the majority were mixed minerals and loose aggregates. Such particles would have lower densities than the bulk material. Many of the clays contained sulfur, which was probably in the form of sulfate on the particles' surfaces. This phenomenon is described in more detail elsewhere.[6] (c) In the samples analyzed, about 25 percent of the coarse particles were of anthropogenic origin and were apportioned as follows: 9 percent were fly ash enriched in sulfur, 11 percent were sulfate particles, and 5 percent contained specific metals that indicated aluminum and iron foundries and incinerators.

Ultrafine Particles

Figure 8 is a micrograph of particles on the chloroform-treated Nuclepore filters. It shows an abundance of chain agglomerates of particles smaller than 0.1 μm. We could not detect trace elements, but because they are so small we cannot exclude the remote possibility that the agglomerates might have resulted from fumes from some metalurgical source. However, we consider it more likely that the chain agglomerates consist of elemental carbon.

Summary and Conclusions

Electron microscopy analysis of particles collected at DCL in western Maryland revealed a relatively limited number of particle categories. Sulfate particles dominate particles in the submicrometer size range. The percentage was higher than in most analyses performed for rural sites in the west and southwest United States[20] or urban sites elsewhere (Y. Mamane, unpublished data). The sulfates were acidic ammonium sulfates such as ammonium bisulfate. Since most sulfates did not contain any large "impurities," and because they were not emitted as large primary sulfate particles, one could suggest a mechanism of homogeneous reaction, probably photochemical, as the main pathway of secondary sulfate formation. Sulfates were also associated with minerals and fly ash particles in the coarse mode, but sulfate comprised a small fraction of the coarse fraction mass.

Nitrates were absent in the 0.3- to 1-μm size range and amount to a few percent of the 0.8- to 2-μm diameter particles. They occurred mainly as mixed nitrates.

Particles larger than 1.5 to 2 μm were completely different from the smaller particles in composition, structure, and homogeneity. Clay minerals, quartz, gypsum, and calcite comprised 50 percent of the coarse particles; spores, pollen, and plant debris comprised about 25 percent. The latter group seems to be highly enriched in sulfur; this suggests the deposition of sulfur dioxide on the pollen surfaces. The rest of the coarse mode particles were of anthropogenic origin, mainly fly ash (~9 percent), sulfates (~11 percent), and few metal particles from upwind smelters or incinerators.

A large portion of the ultra-fine mode chain agglomerates were presumed to be elemental carbon. Only a small fraction of the sulfate particles contained a core of such particles.

Electron microscopy analyses provide information on the size, morphology, surface properties, chemical composition, and heterogeneity of individual particles. This information is crucial in the determination of the sources, transport, and transformation of the particles; thus, electron microscopy is an important tool to be used in receptor modeling to enhance source resolution.

Acknowledgment

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Although research described in this article has been conducted at the U.S. Environmental Protection Agency, it has not been subjected to Agency review and therefore does not necessarily reflect the views of the Agency, and no official endorsement should be inferred. Mention of commercial products and company names does not constitute endorsement by the U.S. EPA.

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References

1. R. K. Stevens, U.S. Environmental Protection Agency, Research Triangle Park, NC, private communication, 1983.
2. R. K. Stevens, T. G. Pace, "Overview of the mathematical and empirical receptor models workshop (Quail Roost II)," *Atmos. Environ.* 18: 1499 (1984).
3. G. E. Gordon, W. R. Pierson, J. M. Daisey, P. J. Lioy, J. A. Cooper, J. G. Watson, G. R. Cass, "Considerations for design of source apportionment studies," *Atmos. Environ.* 18: 1567 (1984).
4. G. S. Casuccio, P. B. Janocko, R. J. Lee, J. F. Kelly, S. L. Dattner, J. S. Mypbroff, "The use of computer controlled scanning electron microscopy in environmental studies," *JAPCA* 33: 937 (1983).
5. D. L. Johnson, B. L. Davis, T. G. Dzubay, H. Hasan, E. R. Crutcher, W. J. Courtney, J. M. Jaklevic, A. C. Thompson, P. K. Hopke, "Chemical and physical analyses of Houston aerosol for interlaboratory comparison of source apportionment procedures," *Atmos.*

Environ. **18:** 1539 (1984).
6. Y. Mamane, K. Noll, "Characterization of large particles at a rural site in the Eastern United States: mass distribution and individual particle analysis," *Atmos. Environ.* **19:** 611 (1985).
7. K. R. May, "The cascade impactor: an instrument for sampling coarse aerosols," *J. Sci. Instrum.* **22:** 187 (1945).
8. Y. Mamane, R. G. de Pena, "A quantitative method for the detection of individual submicrometer size sulfate particles," *Atmos. Environ.* **12:** 69 (1978).
9. Y. Mamane, R. F. Pueschel, "A method for the detection of individual nitrate particles," *Atmos. Environ.* **14:** 629 (1980).
10. B. Y. H. Liu, K. W. Lee, "Efficiency of membrane and Nucleopore filters for submicrometer aerosols," *Environ. Sci. Technol.* **10:** 345 (1976).
11. E. J. Chatfield, R. W. Glass, M. J. Dillon, "Preparation of Water Samples for Asbestos Fiber Counting by Electron Microscopy," EPA-600/4-78-011, 132 pp., Office of Research and Development, U.S. Environmental Protection Agency, Athens, GA, 1978.
12. P. J. Samson, M. E. Ferman, J. M. Masters, D. A. Johnson, "Meteorological Analysis of the 1983 Deep Creek, Maryland Experiment Data," (draft), Department of Atmospheric and Oceanic Science, University of Michigan, Ann Arbor, 1984.
13. Y. Mamane, "A Quantitative Method for the Detection of Individual Submicron Sulfate Particle," Ph.D. Thesis, Pennsylvania State University, University Park, PA 1977.
14. M. J. Heard, R. D. Wiffen, "Electron microscopy of natural aerosols and the identification of particulate ammonium sulphate," *Atmos. Environ.* **3:** 337 (1969).
15. E. R. Frank, J. P. Lodge, "Morphological identification of airborne particles with the electron microscope," *J. Microscopie* **9:** 735 (1967).
16. W. R. Pierson, W. W. Brachaczek, T. J. Truex, J. W. Butler, T. J. Korniski, "Ambient sulfate measurements on Allegheny Mountain and the question of atmospheric sulfate in the northeastern United States," *Ann. N. Y. Acad. Sci.* **338:** 145 (1980).
17. R. W. Shaw, R. J. Paur, "Composition of aerosol particles collected at rural sites in the Ohio River Valley," *Atmos. Environ.* **17:** 2031 (1983).
18. R. K. Stevens, T. G. Dzubay, C. W. Lewis, R. W. Shaw, "Source apportionment methods applied to the determination of the origin of ambient aerosols that affect visibility in forested areas," *Atmos. Environ.* **18:** 261 (1984).
19. K. E. Noll, "A rotary inertial impactor for sampling giant particles in the atmosphere," *Atmos. environ.* **4:** 9 (1970).
20. Y. Mamane, R. F. Pueschel, "On the properties of the condensation nuclei in rural environments in the Western United States," *J. Rech. Atmos.* **14:** 223 (1980).

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